Singlet Excitations in Pyrochlore: A Study of Quantum Frustration

Erez Berg, Ehud Altman and Assa Auerbach
Department of Physics, Technion, Haifa 32000, Israel.

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We apply the Contractor Renormalization (CORE) method to the spin half Heisenberg antiferromagnet on the frustrated checkerboard and pyrochlore lattices. Their ground states are spin-gapped singlets which break lattice symmetry. Their effective Hamiltonians describe fluctuations of orthogonal singlet pairs on tetrahedral blocks, at an emergent low energy scale. We discuss low temperature thermodynamics and new interpretations of finite size numerical data. We argue that our results are common to many models of quantum frustration.

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Frustration in classical spin models often leads to a complex energy landscape. Certain models, such as the Heisenberg antiferromagnet on the pyrochlore lattice, has an extensively degenerate ground state manifold. This model given by

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \tag{1}$$

has spins $\mathbf{S}_i$ sitting on corner sharing tetrahedral units (see Fig. 1(a)).

In the *semiclassical* approximation[1, 2, 3], a large degeneracy survives the quantum fluctuations, and thus resists ground state selection by the "order from disorder" mechanism.

A pressing open question is what happens in the *strong* quantum limit, e.g. the spin half case? Series expansions[5] suggest rapid decay of spin correlations. Does this indicate the formation of a translationally invariant spin liquid or lattice symmetry breaking valence bond solid? In the quantum case, is there an emerging low energy scale, in lieu of the classical ground state degeneracy?

The purpose of this Letter is to derive the low energy effective Hamiltonian starting from the Heisenberg model. As a warm-up to the pyrochlore lattice (Fig. 1(a)) , we treat its two dimensional (toy model) reduction, the Checkerboard lattice (Fig. 1(b)). The Checkerboard has recently received significant theoretical attention[6, 7, 8, 9, 10].

Our approach is the Contractor Renormalization (CORE) method[11]. The CORE is a real-space discrete renormalization transformation invented by Morningstar and Weinstein. It maps a lattice Hamiltonian to an effective Hamiltonian with the same low energy spectrum. The CORE method computes the effective interactions at all ranges using exact diagonalizations of finite connected clusters. Truncation of interactions beyond a finite range is an approximation whose error can be estimated numerically from the next higher range terms. CORE has been successfully applied to describe the spectra of Heisenberg models on chains and ladders[11, 12]. Recently, it was applied to the square lattice Hubbard model to derive the Plaquette Boson-Fermion Model for cuprate superconductors[13]. We refer the reader to previous reviews[11, 13] for the mathematical background and technical details.

For each of the Hamiltonians at hand, we define local operators from the lowest eigenstates of the elementary clusters, e.g. a tetrahedral unit in the Pyrochlore lattice. We shall compute the effective interactions by CORE up to four clusters range, and estimate the truncation error.

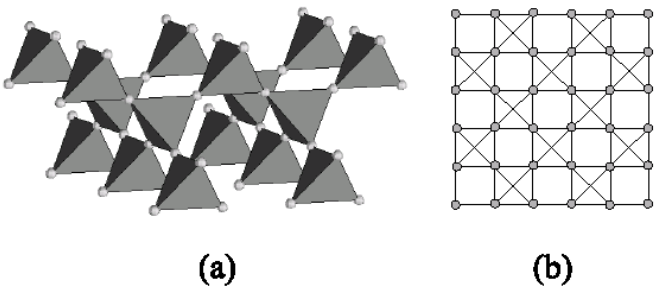

FIG. 1: The Pyrochlore (a) and Checkerboard (b) lattices.

Our key results are the following: *For the Checkerboard,* we confirm the conclusions of refs [9, 10], derived by other means, that the ground state is closely approximated by the product of uncrossed plaquette singlets. The effective pseudospin Hamiltonian allows us to interpret the numerical spectrum of low lying singlets[6, 7] in terms of Ising domain walls. This yields the number of singlets as a function of lattice size: a numerically testable prediction. In addition we expect a branch of weakly dispersive triplets at an energy scale slightly below the Heisenberg exchange. *For the Pyrochlore,* we apply two CORE steps to arrive at an Ising-like model of local singlets on larger super-tetrahedra blocks which form a cubic superlattice. At mean field level, we obtain a singlet ground state which breaks lattice symmetry as depicted in Fig.5. Here too, the effective Hamiltonian

describes Ising-like domain walls. We shall conclude that lattice symmetry breaking and local singlet excitations are general features of highly frustrated quantum antiferromagnets.

The Checkerboard. The lattice depicted in Fig. 1(b), contains crossed plaquettes (which are equivalent to three dimensional tetrahedra) connected by uncrossed plaquettes. The first step is to choose elementary clusters which cover the lattice. Two different options for plaquettes are the *crossed* and the *uncrossed plaquettes*. While the two clustering choices may appear to yield different ground states and excitations, we shall see that they are in fact consistent with each other, and yield complementary information.

Clustering with uncrossed plaquettes. From the spectrum of a single uncrossed plaquette, we retain the singlet ground state as a vacuum state $|\,\Omega\,\rangle_i$ and the lowest triplet as a singly occupied boson state $t^\dagger_{\alpha i}\,|\,\Omega\,\rangle_i$. $\alpha = x, y, z$ is a cartesian index of the triplet.

The effective Hamiltonian in the uncrossed plaquettes basis is (in units of $J = 1$):

$$\mathcal{H}_{eff} = \epsilon'_t \sum_i t^\dagger_{\alpha i} t_{\alpha i} + K \sum_{\langle\, i\, j\,\rangle} \mathbf{L}_i \cdot \mathbf{L}_j + \sum_{\langle\, i\, j\,\rangle\, \alpha\beta} \{-\Delta t^\dagger_{\alpha i} t^\dagger_{\alpha j} t_{\beta i} t_{\beta j} + b t^\dagger_{\alpha i} t_{\alpha i} t^\dagger_{\beta j} t_{\beta j}\}, \tag{2}$$

where $\mathbf{L}_j = \sum_{\alpha\beta} t^\dagger_{\alpha j} \vec{\mathcal{L}}_{\alpha\beta} t_{\beta j}$ and $\vec{\mathcal{L}}_{\alpha\beta}$ are 3×3 spin-1 matrices in a cartesian basis.

The parameters calculated by CORE upto range 2 are: $\epsilon'_t = 0.5940$, $K = 0.2985$, $\Delta = 0.1656$ and $b = 0.0776$. The truncation errors from up to range-4 are less than 2%, and will be ignored[14].

Note that $\mathcal{H}_{eff}$ in (2) commutes with the number of triplets since it has no anomalous pair creation terms, as appear e.g. for the square lattice [13]. Thus, at this level of truncation, the plaquette vacua product

$$|\Psi_0\rangle = \prod_i |\Omega_i\rangle, \tag{3}$$

is an exact ground state of the effective Hamiltonian (2)[15]. This result agrees with Moessner *et al* [9], who argued for a plaquettized singlet ground state based on an effective quantum dimer model.

We are also able to obtain the triplet (spin) gap for Since $t^\dagger_{\alpha,i}|0\rangle$ is an approximate eigenstate of (2), its energy (spin gap) can be read from $\epsilon'_t = 0.5940$. This compares well with the value of 0.6-0.7 estimated by exact diagonalizations of finite systems [7]. We have found very weak hopping terms (of magnitude $0.01J$) due to CORE interactions of range four, which will give the triplets a weak dispersion in the full lattice.

Clustering with crossed plaquettes. The isolated crossed plaquette has two fold degenerate singlet ground states, which we can represent by a pseudospin-$\frac{1}{2}$ doublet (see Fig. 2).

The quantization axis for the pseudospin operators is chosen as in Ref. [16], with the $+z(-z)$ directions representing states with positive (negative) chirality. The planar angles $0, \pi/3, 2\pi/3$, represent the three (non orthogonal) dimer configurations of the tetrahedron. The states with their pseudospin polarized in the $+x$ and $-x$ direction are shown in Fig. 2.

The effective Hamiltonian in the crossed plaquettes basis is an Ising-like model

$$H_{eff} = -J' \sum_{\langle ij\rangle} (\mathbf{S}_i \cdot \mathbf{e}_{ij})(\mathbf{S}_j \cdot \mathbf{e}_{ij}) - h \sum_i S^x_i. \tag{4}$$

where $\mathbf{e}_{ij}$ are directors on the $x - y$ plane pointing $\pi/3$ $(-\pi/3)$ away from the x axis for horizontal (vertical) bonds. At range-2 we obtain, $J' = 0.527665$, and $, h = 0.118084$ Corrections from range-3 and range-4 CORE were computed[14], and found to be unimportant for the symmetry and correlations of the low excitations.

We can solve (4) in mean field theory. The energy exhibits two minima, where the pseudospins describe vertical or horizontal dimers. These states correspond respectively to projections of the two equivalent plaquette ground states onto the truncated Hilbert space of the crossed plaquettes. Although the ground state energy is not well converged at range-2 CORE, (4) treats the two symmetry breaking ground states in an unbiased fashion. It therefore describes low energy singlet excitations which are pseudospin-flips or Ising domain walls between ground states (see Fig. 3).

Thus we see that the two methods elucidate complimentary aspects of the Checkerboard. The approach using uncrossed plaquettes gives a very accurate description of one ground state, which serves as its vacuum, but where the other ground state is a multi-magnon bound state. In contrast, the crossed plaquettes describes correctly the low energy singlet excitations by an effective Ising model $J^{Ising} S^z S^z$ with a coupling constant $J^{Ising} = 9/8 J'$ renormalized by the quantum fluctuations. The lowest excitations are gapped spin flips of energy J^{Ising}. They gain a weak dispersion due to the effective $S^+ S^-$ couplings.

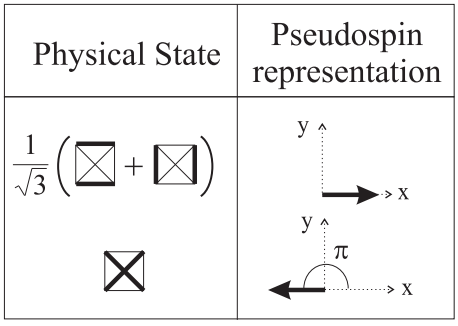

FIG. 2: The two singlet ground states of the uncrossed plaquette and their pseudospin representation. In the physical state thick lines denote valence bond singlets.

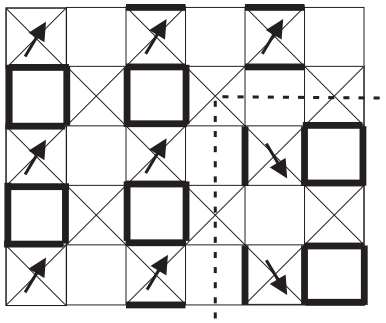

FIG. 3: A singlet excitation of the checkerboard. This state is a domain wall (dashed line) between domains of the two plaquettized ground states. The corresponding pseudospin directions in the crossed plaquette approach are depicted by arrows.

The number of singlet states is expected to grow as *power laws* with the size of the system N. For example,the lowest lying single spin-flips grow as N, while higher spin flip pairs grow as N^2 and so on. An Ising-like phase transition is expected between the broken and unbroken translational symmetry phases at a temperature scale of $T_c \simeq J^{Ising}$, with an associated logarithmic divergence of the heat capacity at T_c.

The Pyrochlore, depicted in Fig. 1(a), is a three dimensional network of corner sharing tetrahedra. Like the checkerboard, it has a macroscopically degenerate (exponential in lattice size) classical ground state manifold. For the quantum S=1/2 case, local pseudospins can be defined by the degenerate singlets of disjoint tetrahedra. These cover all sites of the Pyrochlore and form an FCC superlattice. The effective hamiltonian on this FCC lattice was calculated by CORE. The first non-trivial inter-teterahedra coupling are obtained at range three connected tetrahedra, which yield

$$H^{(3)}_{eff} = \sum_{\langle ijk \rangle} \Big((J_2(\mathbf{S}_i \cdot \mathbf{e}^{(i)}_{ijk})(\mathbf{S}_j \cdot \mathbf{e}^{(j)}_{ijk}) + J_3(\frac{1}{2} - \mathbf{S}_i \cdot \mathbf{e}^{(i)}_{ijk})(\frac{1}{2} - \mathbf{S}_j \cdot \mathbf{e}^{(j)}_{ijk})(\frac{1}{2} - \mathbf{S}_k \cdot \mathbf{e}^{(k)}_{ijk}) \Big). \quad (5)$$

The coupling parameters (in units of J) are: $J_2 = 0.1049$, $J_3 = 0.4215$, and $\mathbf{e}^{(i)}_{123}, i = 1,2,3$ are three unit vectors in the x-y plane whose angles $\alpha^{(i)}_{123}$ depend on the particular plane defined by the triangle of tetrahedral units 123 as given in table I of [16]. The effective hamiltonian (5) resembles the terms obtained by second order perturbation theory (in inter-tetrahedra couplings) by Harris *et al* [4] and Tsunetugu[16]. The classical mean field ground state of (5) is identical to the ground state found in Ref.[16]: three of the four FCC sublattices are ordered in the directions $\mathbf{e}(0), \mathbf{e}(2\pi/3), \mathbf{e}(-2\pi/3)$, while the direction of the fourth is completely degenerate. Therefore, classical mean field approximation for (5) is insufficient to remove the ground state degeneracy. Tsunetsugu[16] was able to lift the degeneracy by including spinwave fluctuations effects which produce ordering at a new low energy scale.

Here we avoid the *a-priori* symmetry breaking needed for semiclassical spinwave theory, by treating (5) fully quantum mechanically. This entails a second CORE transformation which involves choosing the *"supertetrahedron"*, as a basic cluster of four tetrahedra, whose structure and spectrum are depicted in Fig.4.

Our new pseudospins τ_i are defined by the two degerenate singlet ground states of the supertetrahedron. (This degeneracy is found for the Heisenberg model on the original lattice as well as for the effective model (5)). These states transform as the E irreducible representation of the tetrahedron (T_d) symmetry group, similarly to the singlet ground states of a single tetrahedron.

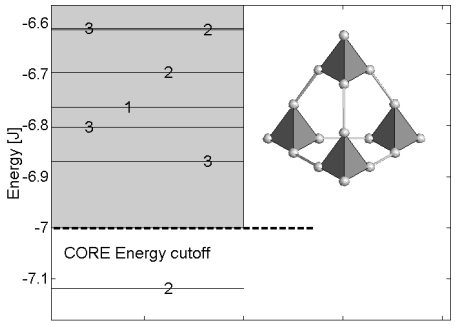

FIG. 4: Low energy spectrum of a single "supertetrahedron". A dashed line illustrates the energy cutoff: only the two degenerate ground states are retained in the Hilbert space.

The supertetrahedra form a cubic lattice, shown in Fig. 5. The effective hamiltonian (5) and the lattice geometry imply that non-trivial effective interactions appear only at the range of three supertetrahedra and higher. Range three effective interactions include two and three pseudospin interactions, which are dominated by

$$\mathcal{H}_{eff} = J_1 \sum_{\langle ij \rangle} (\tau_i \cdot \mathbf{f}_{ij})(\tau_j \cdot \mathbf{f}_{ij}) + J_2^{(a)} \sum_{\langle\langle ij \rangle\rangle} (\tau_i \cdot \mathbf{f}_{ij})(\tau_j \cdot \mathbf{f}_{ij}) + J_2^{(b)} \sum_{\langle\langle ij \rangle\rangle} (\tau_i \cdot (\mathbf{f}_{ij} \times \hat{\mathbf{z}}))(\tau_j \cdot (\mathbf{f}_{ij} \times \hat{\mathbf{z}})). \quad (6)$$

Here, $\langle\ \rangle$ and $\langle\langle\ \rangle\rangle$ indicate summation over nearest- and next nearest-neighbors, respectively. The coupling constants are found to be relatively small: $J_1 = 0.048J$, $J_2^{(a)} = -0.006J$ and $J_2^{(b)} = 0.018J$. The vectors $\mathbf{f}_{ij}$ depend on the vector $\mathbf{r}_{ij}$ connecting the two sites, and their values are presented in table .

We performed classical Monte Carlo simulations using the classical (large spin) approximation to (6). The

TABLE I: The values of the vectors $\mathbf{f}_{ij}$ in eq. (6), depending on the vector $\mathbf{r}_{ij}$ separating the sites i and j.

$\mathbf{r}_{ij}$	$\mathbf{f}_{ij}$
$(\pm1,0,0)$ $(0,\pm1,\pm1)$	$(1,0,0)$
$(0,\pm1,0)$ $(\pm1,0,\pm1)$	$(-\frac{1}{2},\frac{\sqrt{3}}{2},0)$
$(0,0,\pm1)$ $(\pm1,\pm1,0)$	$(-\frac{1}{2},-\frac{\sqrt{3}}{2},0)$

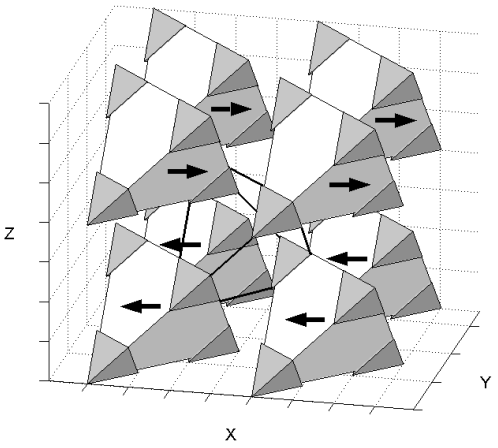

FIG. 5: The pyrochlore viewed as a cubic lattice of supertetrahedra. The arrows show the direction of the supertetrahedra's pseudospin in the MF ground state.

ground state was found to choose an antiferromagnetic axis, and to be ferromagnetic in the planes as depicted in Fig. 5. This ground state differs from the semiclassical ground state[4, 16]. The latter involves condensation of high energy states of the supertetrahedron in the thermodynamic ground state. Since in the supertetrahedra diagonalizations, we find a much larger energy gap than inter-site coupling we believe these excitations cannot condense to yield the semiclassical ground state symmetry breaking.

Discussion. The CORE technique enabled us to derive effective Hamiltonian for highly frustrated antiferromagnets, written in terms of low energy, local degrees of freedom. For both Checkerboard and Pyrochlore systems, we found lattice symmetry breaking ground states which are essentially products of local singlets. The spin gap to the lowest triplet excitation is large (of order J), and seems to survive interplaquette interactions. The low energy excitations are singlets, which are local pseudospin flips, or Ising domain walls between ground states. The ordering energy scale is of order $J/100$ for the Pyrochlore.

This picture seems to be consistent with existing numerical data for the density of low energy singlets on the Checkerboard and Pyrochlore. Experimentally, lattice symmetry breaking could drive a static lattice distortion, which would be observable by additional Bragg peaks in neutron and X-ray scattering. For example, the antiferromagnetic order between planes of supertetrahedra would correspond to a lattice distortion with wavelength of four tetrahedra.

How general are these results? Formation of local singlets is a natural way to relieve the frustration in quantum antiferromagnets that can be written as a sum over clusters $\sum_c(\sum_i \mathbf{S}_{ic})^2$. On each even cluster the ground state is a singlet with a gap of order $1/S$ to a local triplet. Frustration suppresses hopping of these triplets and could inhibit their condensation into a spin ordered ground state. Thus lattice symmetry breaking singlet ground states are expected as a typical feature of frustrated quantum antiferromagnets[17].

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